

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Re: Application of LALANNE-MAGNE et al.

Serial No: 10/509,884

Filed: October 4th 2004

For: PROCESS FOR THE GAS PHASE (CO-)POLYMERISATION OF OLEFINS
IN A FLUIDISED BED REACTOR

DECLARATION OF CLAUDINE LALANNE-MAGNE

I, CLAUDINE LALANNE-MAGNE, do hereby solemnly declare as follows:-

1. I am a French subject, residing at 32 bis Bd Pablo Neruda-13920 St Mitre les Remparts- France
2. I graduated from ENSIC Nancy, in France in 1985 with a Chemical Engineer Degree.
3. I am currently employed by INEOS Technologies at the INEOS Research and Technology Centre, Lavéra, France. I am currently Gas Phase PE Development Manager and worked in several other eras such as pilot plant, manufacturing, licensing and plant support activities all in the PE gas phase area for almost 23 years. Since 1985, I have been working on the Innovene (formerly BP) fluidised bed process for manufacture of polyethylene.
4. I am familiar with the technical details concerning the US Patent Application Serial No: 10/509,884 filed on October 4th 2004, of which I am an inventor.
5. I have read and am familiar with the cited art, US 5,990,251 to Gelus. In fact, I was an employee of BP in the Research and Technology Centre in Lavera, France from which this cited patent originates at the time of its filing, my current employee, INEOS Technologies, being the successor of BP at the Research and Technology Centre in Lavera.
6. The present invention relates to a process for the gas-phase (co-)polymerisation of olefins in a fluidized bed reactor using a Ziegler-Natta type catalyst, said process comprising the addition into the reactor of an organoaluminium cocatalyst and of a monohalogenated hydrocarbon compound, wherein the molar ratio of the monohalogenated hydrocarbon compound to the cocatalyst is between 0.02 and 0.2, the monohalogenated hydrocarbon compound is added to the reactor in an amount comprised between 0.625 and 40 moles of monohalogenated hydrocarbon compound per mole of transition metal of catalyst introduced into the reactor, and wherein the monohalogenated hydrocarbon compound is n-butyl chloride.

7. Gelus also relates to a process for polymerizing olefin(s) in the presence of a catalyst of Ziegler-Natta type and, in particular, to the use of halogenated hydrocarbon compounds as catalyst activating agents.
8. Gelus lists a number of potential halogenated hydrocarbon compounds that can be used as activators. However, n-butyl chloride is not explicitly mentioned, and the only halogenated hydrocarbon compound actually exemplified in Gelus is chloroform.
9. Further, Gelus discloses that there should be used a halogenated hydrocarbon to catalyst transition metal molar ratio at its broadest of 0.001 to 0.15 (col. 2, lines 42-47). In contrast, the present invention requires a ratio of at least 0.625.
10. In further support of the patentability of the present invention, the attached Appendix 1 presents data comparing n-butyl chloride to chloroform and a number of similar monohalogenated hydrocarbons. Under the conditions used, the Appendix shows that n-butyl chloride provides a significant enhancement in activity (53%) compared to the unpromoted system. This enhancement is significantly higher than that observed for the other monoalkylchlorides tested, some of which actually reduce the activity.
11. Under the conditions of Appendix 1, chloroform reduces the activity compared to an unpromoted system. Further, the enhancement seen for n-BuCl under these conditions is higher than the maximum increase (approximately 33%) shown for the use of chloroform in Figure 2 of Gelus. Thus, the relative increase in activity by use of n-BuCl shown in Appendix 1 is also higher than the maximum increase taught by Gelus even when using chloroform in the range taught by Gelus.
12. Not least for the above reasons, it is believed that the selection of n-BuCl and its use in the amount relative to titanium as claimed in the present claims is not obvious over the cited art.

All statements made of my own knowledge are true and all statements made on information and belief are believed to be true. I also understand that wilful false statements and the like are punishable by fine or imprisonment, or both (18 U.S.C. 1001) and may jeopardize the validity of the application or any patent issuing thereon.

CLAUDINE LALANNE-MAGNE

Date: 22/4/05

Claudine Lalanne-Magne

Appendix 1

A number of potential activity promoters were tested in a 2,5L gas phase reactor following an equivalent procedure in each case. For each reaction, the ethylene pressure was fixed at 6 bars and 2.5 bars of hydrogen were added for the molecular weight control. The polymerisation temperature was set to 90°C. The quantity of the halogenated compounds was such as to target a molar ratio of the halogenated compound to Ti of 40:1.

The catalyst used was a Ziegler-Natta catalyst similar to that used in the Examples of the present invention.

The results are shown in Figure 1, along with comparison with a reference sample in which no activity promoter was used.

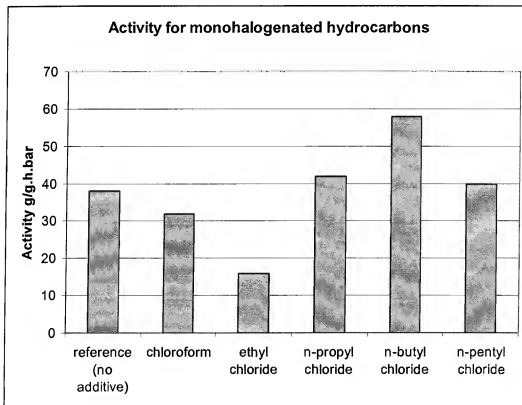


Figure 1 : Activity data for a series of monohalogenated hydrocarbons.

As can be seen from the above Figure, n-butylchloride (n-BuCl) provides a significant enhancement in activity (53%) compared to the unpromoted system.

(Please note that the activity above is quoted as g/g.hour.bar (grams of polymer produced per gram of titanium per hour per bar) – under the conditions of these reactions this is about a factor of 10 less than the equivalent values quoted as g/mmol Ti per hour per 0.1MPa as used in the present application.)